Direct contact between dust and HBCD-treated fabrics is an important pathway of source-to-dust transfer

Rauert, Cassandra; Kuribara, Isamu; Kataoka, Toshiyuki; Wada, Takeharu; Kajiwara, Natsuko; Suzuki, Go; Takigami, Hidetaka; Harrad, Stuart

DOI:
10.1016/j.scitotenv.2015.12.054

Document Version
Peer reviewed version

Citation for published version (Harvard):
Rauert, C, Kuribara, I, Kataoka, T, Wada, T, Kajiwara, N, Suzuki, G, Takigami, H & Harrad, S 2016, 'Direct contact between dust and HBCD-treated fabrics is an important pathway of source-to-dust transfer', *Science of the Total Environment*, vol. 545-546, pp. 77-83. https://doi.org/10.1016/j.scitotenv.2015.12.054

DIRECT CONTACT BETWEEN DUST AND HBCD-TREATED FABRICS IS AN IMPORTANT PATHWAY OF SOURCE-TO-DUST TRANSFER

Cassandra Rauert[a], Isamu Kuribara[b], Toshiyuki Kataoka[b], Takeharu Wada[b], Natsuko Kajiwara[c], Go Suzuki[c], Hidetaka Takigami[c], Stuart Harrad[a]

[a] School of Geography, Earth, and Environmental Sciences, University of Birmingham, Birmingham, B15 2TT, UK

[b] Chemicals Evaluation and Research Institute, Japan (CERI), 1600 Shimotakano, Sugito-machi, 345-0043, Japan

[c] Center for Material Cycles and Waste Management Research, National Institute for Environmental Studies (NIES), 16-2 Onogawa, Tsukuba, 305-8506, Japan

Corresponding Author

Stuart Harrad

Tel +44 (0)121 414 7298

S.J.Harrad@bham.ac.uk

Abstract

Hexabromocyclododecanes (HBCDs) are a class of brominated flame retardant that have found extensive application in consumer products used widely in indoor environments. Although uncertainty remains about the human health impacts of HBCDs, ingestion of HBCD-contaminated indoor dust has been shown to be a particularly significant exposure pathway for young children. Despite this, understanding of the mechanisms via which HBCDs transfer from products to indoor dust remains incomplete. In this study, an in-house test chamber was used to investigate transfer of HBCDs from a treated textile sample to indoor dust via direct textile:dust contact. Results were compared with previous data using the same test chamber to examine other pathways via which HBCDs transfer from products to dust, and highlighted HBCD transfer *via* direct source:dust contact as being particularly important. This novel finding was corroborated by complementary experiments that examined HBCD transfer via direct contact, from other treated textiles to three major components of indoor dust: artificial indoor dust, soil particles, and cotton linters.

Keywords

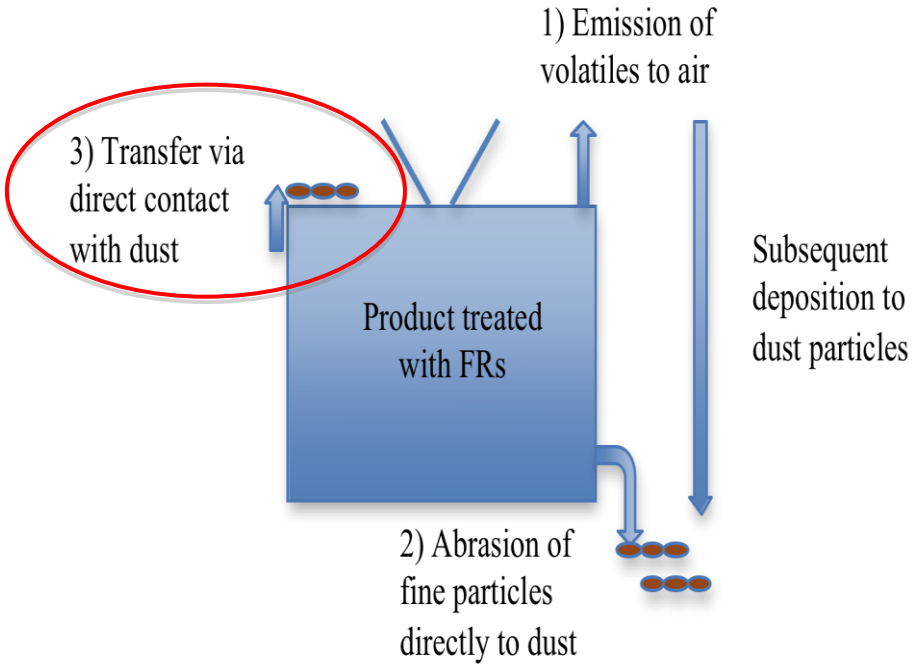
Brominated flame retardants;

Indoor dust;

Source-to-dust Transfer;

HBCD

Graphical abstract



Highlights

Transfer of HBCDs via direct contact between a curtain and dust studied.

Direct curtain:dust contact led to substantial transfer.

Transfer is rapid yet source:dust equilibrium not reached after 1 week of contact.

Results imply regular cleaning of source surfaces may reduce contamination of dust.

Introduction

Hexabromocyclododecanes (HBCDs) are one of the most widely produced classes of brominated flame retardants (BFRs) used to flame retard polystyrene foams for building insulation, fabrics like furniture covers and curtains, and high impact polystyrene casings for electronic equipment (Harrad et al, 2010; Weil and Levchik, 2007). They are incorporated into products *via* an "additive" process where the HBCD formulation is physically rather than chemically bound to the product/polymer. Consequently, their release into the surrounding environment is relatively facile, leading to their ubiquitous presence in indoor air and dust (Harrad et al, 2010; Covaci et al, 2006). Concentrations in dust can vary by orders of magnitude (Abdallah et al, 2008b) up to 1.1 mg ΣHBCDs g^{-1} (Allen et al, 2013). Such elevated concentrations are of concern, given dust ingestion contributes an estimated 63% of the exposure of UK toddlers to ΣHBCDs (Abdallah et al, 2008a).

Currently hypothesized pathways of BFR transfer from products to dust include: (1) volatilization of BFRs from products with subsequent partitioning to dust; (2) abrasion via physical wear and tear of products, resulting in transfer of particles or fibers of the product directly to dust and (3) transfer via direct contact between product and dust (Suzuki et al, 2009; Takigami et al, 2008; Wagner et al, 2013; Webster et al, 2009). Test chambers constitute a potentially important tool for investigating source-to-dust transfer of BFRs. While test chamber studies to date have focused largely on contaminant emissions to air to determine specific emission rates (SERs) (Rauert et al, 2014a), the mass transfer to dust of phthalates from wall paint and vinyl flooring has been investigated in modified test chambers (Clausen et al, 2004; Schripp et al, 2010). These studies demonstrated phthalate transfer from source

to dust occurred *via* volatilization with subsequent partitioning to dust, and *via* direct source:dust contact. More recently, a test chamber study by two of the current authors demonstrated rapid and substantial transfer of PBDEs from a TV casing sample to dust (Rauert and Harrad, 2015).

We have reported previously test chamber experiments simulating source-to-dust transfer of HBCD *via* partitioning post volatilization and *via* abrasion (Rauert et al, 2014b, 2015). However, to our knowledge, transfer of HBCD via direct source:dust contact has hitherto not been investigated.

This study is the first experimental investigation of HBCD transfer to dust through direct source:dust contact, using a test chamber and a HBCD-treated curtain as the source. Results are compared to previous data reporting HBCD transfer to dust *via* transfer pathways (1) and (2). Moreover, given the lack of previous data concerning transfer of HBCDs via direct source:dust contact; we report a series of complementary, more detailed follow-up experiments that examine HBCD transfer via direct contact, from four different treated textiles to three major components of indoor dust: artificial indoor dust, soil particles, and cotton linters.

Materials and Methods

Experimental Design

Initial Test Chamber Experiments

The cylindrical stainless steel test chamber employed to investigate HBCD migration from the test curtain to dust is illustrated in Figure 1a. With dimensions of 10 cm diameter and 20 cm height, volume of 1570 cm^3, and internal surface area of 785 cm^2,

chamber internal temperature was monitored by a LogTag TRIX-8 temperature data logger. An aluminium mesh shelf was placed half way down the chamber. A HBCD treated curtain (7 cm x 3 cm rectangle) was placed on a clean glass fiber filter situated on the shelf, and a thin layer of previously characterised house dust (Rauert et al, 2015) placed evenly on top of the curtain (~500 mg). Concentrations of HBCD diastereomers in the curtains used in test chamber experiments were 18,000,000 ng g^{-1} for α-HBCD, 7,500,000 ng g^{-1} for β-HBCD, and 17,000,000 ng g^{-1} for γ-HBCD (Kajiwara et al, 2013). Dust utilized in test chamber experiments contained low concentrations of HBCDs and PBDEs (ΣHBCDs = 110 ng g^{-1} and ΣPBDEs = 280 ng g^{-1}). The percentage carbon and nitrogen content of this dust was determined, with results supplied as supplementary data (Table SD-1).

The chamber was sealed and left at room temperature (22 ± 1 °C) for either 24 hours or 1 week. Post-experiment dust was gently removed from the source by gentle tapping, and homogenized via vortex mixing, ready for analysis. Each time period was studied in quadruplicate with duplicate dust subsamples (200 mg) from each experiment analyzed for HBCDs.

Follow-up experiments

Figure 1b illustrates the experimental configuration employed in follow-up experiments. Four different kinds of HBCD-treated fabrics including 2 curtains (Curtain-1 and -2) and 2 vehicle seat fabrics (Seat fabric-1 and -2) were used. These materials were manufactured in Japan prior to HBCD's listing as a persistent organic pollutant (POP) under the Stockholm Convention (Annex A). HBCD concentrations in these fabrics ranged from 8,400,000 ng g^{-1} to 21,000,000 ng g^{-1} (Table 3). Each

fabric material tested was fixed within a rectangular aluminium frame (outer dimensions 115 mm x 150 mm) with two windows each 53 mm x 83 mm. Prior to each experiment, the test fabric surface was vacuumed to remove small abraded fibers to minimize their inadvertent incorporation in the test dust. Three different types of test particles were used in follow-up experiments. These were: (1) artificial indoor dust (JIS Z 8901 Class 15, see below); (2) fine soil particle (calcined Kanto loam at 800 ˚C, JIS Z 8901 Class 8, median diameter 6.6–8.6 μm, density 2.9–3.1 g/cm^3, ignition loss 0–4%); and (3) cotton linters (<1.5 μm in diameter, <1 mm in length) (JIS, 2006). The artificial indoor dust was a mixture of test particles, containing 72% fine soil particles (JIS Z 8901 Class 8), 23% carbon black (JIS Z 8901 Class 12, 0.03–0.20 μm, 1.7–1.9 g/cm^3) and 5% cotton linters. The fine soil particles used are primarily inorganic, with negligible organic matter since they were prepared by calcination of loam soil at 800 ˚C. Initial concentrations of HBCDs in the three test dust types were all below detection limits.

Approximately 20-50 mg of test dust, soil or lint were placed within a 25 mm diameter circle on the surface of the HBCD-treated fabric material in the upper window (Figure 1b) using a glass cylinder. Excessive dust was removed by gently tapping the reverse side of the aluminium frame. The exposed fabric surface in the lower window (Figure 1b) was used as a control without dust. An additional blank using dust applied to aluminium foil, instead of a HBCD treated fabric, was also conducted. Each frame was then shielded from light with aluminium foil and maintained at 28˚C and 50% relative humidity in a thermohygrostat test chamber (IG400, Yamato Scientific, Tokyo, Japan). Three experimental durations were examined: 1, 4, and 7 days. Transfer of HBCDs to artificial indoor dust was examined

for each of the 4 HBCD-treated fabric materials over three experimental durations. In contrast, transfer to the fine soil particle and cotton linter samples were examined over a single 4 day period for curtains only. Duplicate tests were conducted for Curtain-1 (all dust types) and Seat fabric-1 (artificial indoor dust only). Following each experiment, dust was collected from the surface of each fabric window (with or without dust) and from the aluminium foil by a low volume vacuum pump equipped with a stainless steel filter holder (KS-25, ADVANTEC, Tokyo, Japan) containing a glass fiber filter (GB-100R, 25 cm diameter, 0.6 μm pore size, ADVANTEC, Tokyo, Japan). Dust collected was weighed and analyzed for HBCDs.

Determination of concentrations of HBCDs

Test chamber experiment samples

Dust from test chamber experiments was analyzed using modified in-house methods (Rauert et al, 2015). A detailed description is provided as supplementary data. Method blanks run with each batch of samples were conducted by extracting a pre-cleaned 66 mL cell filled with hydromatrix. For ^{13}C-α-, β-, and γ-HBCDs, average recoveries ranged from 71 to 85%. Accuracy and precision was assessed via replicate analyses (n=15) of NIST SRM 2585. Results were compared with indicative values (supplementary information, Table SD-2) (Keller et al, 2007). Statistical analysis was conducted using SPSS Version 22 with significance levels set at 0.05.

Follow-up experiment samples

A full description of the methods used to determine HBCDs in follow-up experiments is supplied as supplementary data. Method blanks in follow-up experiments were conducted in parallel via two types of blank test: (1) a dust blank using aluminium foil

instead of HBCD treated fabric to examine cross contamination between test samples; and (2) a fabric blank test whereby the fabric was vacuumed in the absence of dust to evaluate possible co-removal onto the glass fiber filter of small fabric fibers during vacuuming. Concentrations of HBCDs in the dust blank were below detection limits. In fabric blank samples, concentrations of HBCDs were about 100 times lower than those obtained in the presence of dust.

Results and Discussion

Test chamber experiments examining HBCD transfer via direct contact between source:dust

Average concentrations of HBCDs in dust (ng g^{-1}) from pre- and post-chamber experiments are given in Table 1. Regardless of duration, a clear increase in HBCD dust concentrations was observed post-experiment. However, no statistically significant difference ($p>0.1$, independent samples t-test) was observed between concentrations after 24 hours (1,500-14,000 ng g^{-1} ΣHBCDs) and 1 week's contact (5,200-9,300 ng g^{-1} ΣHBCDs).

However, replicate chamber experiments conducted over the same experimental duration displayed substantial (up to an order of magnitude) differences in concentrations of HBCDs in dust post-experiment. Moreover, HBCD concentrations varied considerably between sub-samples of dust taken at the end of the same experiment. This sizeable intra-experimental variation in HBCD concentrations in post-experiment dust made any increase in HBCD transfer between the 24 hour and 1 week experiments difficult to discern. Hence, these experiments could not evaluate reliably whether source-to-dust transfer occurred rapidly and whether equilibrium was

reached within 1 week. We believe the variable HBCD concentrations in these initial experiments most likely arose from uneven transfer of abraded curtain fibers into dust when sampled post-experiment. At the end of each experiment, dust was collected by gently brushing/tapping the fabric. This may have caused transfer of friable curtain fibers, which if heterogeneously distributed within the dust will cause an inhomogeneous distribution of HBCD throughout the dust. This hypothesis is supported by Clausen et al (2004) who investigated direct transfer of di(2-ethylhexyl) phthalate (DEHP) via contact between treated polyvinylchloride (PVC) pieces and dust. The authors suggested vacuuming the dust from the source surface affected the transfer of abraded PVC particles treated with DEHP into the dust.

Coupled with this, it is possible dust in contact with the curtain will display a heterogeneous distribution of organic carbon. HBCDs sorb more strongly to matrices with higher organic carbon content (Abdallah et al, 2012). In the context of source-to-dust transfer of organic contaminants, a weak correlation between uptake of di-n-butyl phthalate and the organic content of the receiving matrix was reported (Schripp et al, 2010). Consequently, variations in organic carbon distribution throughout the receiving dust could partly account for variable HBCD concentrations in dust post-experiment. To examine this further, the organic carbon content of test chamber experiment dust was determined in quadruplicate, revealing an average organic carbon concentration of 12.7% with a standard deviation of 2.5% (Table SD-1), consistent with a heterogeneous distribution.

Source-to-dust transfer is a function of the fugacity gradient of HBCD in the textile surface and dust, if the source:air interface boundary layer is partially absent (Clausen

et al, 2004). A fugacity gradient will promote HBCD transfer from the material in which HBCD fugacity is high to the material in which its fugacity is lower, and the stronger this fugacity gradient, the more rapid the mass transfer. The fugacity is a product of the fugacity capacity of the material and the HBCD concentration in the material. Hence, the fugacity gradient between the two materials is driven largely by the HBCD concentration gradient. Given HBCD concentrations in the curtain exceed substantially those in pre-experiment dust, transfer should be substantial and rapid, with both observations consistent with those seen here.

While this study is only the second to address source-to-dust transfer of BFRs, others have investigated uptake of phthalates to dust via direct contact, successfully replicating this migration pathway in test cell experiments (Clausen et al, 2004; Schripp et al, 2010). Schripp et al (2010) hypothesized such mass transfer results from contact between the dust mass and the boundary layer of the source:air interface. HBCD concentrations in the boundary layer will exceed substantially those in the well-mixed air above, providing increased contact with and rapid transfer to dust, due to the greater gradient between HBCD concentrations in boundary layer air and the dust. However, Clausen et al (2004) suggested direct contact between dust and source partially eliminates the boundary layer, allowing dust particles to act as a sorbent accumulating the contaminant directly from the source surface. In this scenario, dust uptake will be less dependent on contaminant volatility and more on its availability at the source surface.

Comparison of direct source:dust contact with other source-to-dust transfer pathways

We have reported previously experiments using different configurations of the test chamber used here, that demonstrated migration of HBCDs from the same curtain fabric to dust *via* partitioning post-volatilization and abrasion (Rauert et al, 2014b, 2015). Table 2 compares concentrations of HBCDs in dust following these earlier test chamber experiments with those arising via direct source:dust contact.

Table 2 shows migration *via* direct contact between the HBCD source and dust was rapid and yielded higher HBCD concentrations in dust (1,500-14,000 ng g^{-1} ΣHBCDs) than the volatilization experiments. Consistent with this, Clausen et al (2004) reported increased mass (up to four times) of DEHP transferred to dust *via* direct contact than emitted to air from the source over the same experimental duration. Similarly rapid transfer via direct source:dust contact was observed for BDE-209 (Rauert and Harrad, 2015). Moreover, while Table 2 shows the longer duration abrasion experiments afforded higher HBCD concentrations in receiving dust; concentrations in 2 h and 3 h abrasion experiments were very similar to those from our direct source:dust contact experiments. This is important, given Rauert et al (2014b) reported the experimental durations of the abrasion experiments, likely exceed realistic wear and tear of domestic soft furnishings and curtains. While it is recognised that abrasion of the source is possible in these direct contact experiments and may contribute some mass transfer (and explain heterogeneity of HBCDs in the dust); we do not believe it can entirely account for the large concentration increase observed, due to the gentle handling of the source material (and the absence of any visible fibres in the sampled dust). Table 2 highlights the importance of direct source:dust contact as a pathway via which HBCDs (and by extrapolation, similar contaminants) transfer from source materials to dust.

Results of Follow-up Experiments

Figure 2 gives concentrations of ΣHBCDs in the artificial indoor dust samples analysed, with concentrations of individual HBCD diastereomers supplied as Table 4). While initial concentrations in dust (pre-contact with fabrics) were below detection limits, concentrations after just 1 day's contact exceeded 2,000 ng g^{-1} and reached 23,000 ng g^{-1} in artificial dust in contact with Seat fabric-2 for 7 days. This demonstrates substantial HBCD transfer following direct contact between fabrics and dust, and confirms the findings of the test chamber experiments, that this pathway is a highly effective vector via which HBCDs transfer from treated fabrics to dust.

In test chamber experiments, HBCD concentrations in dust in contact with the curtain for 1 week were not significantly different to those after 24 hours of contact, likely due to between-dust sample variations in inadvertent transfer of curtain fibers during dust sample collection and the organic carbon content of the dust. In follow-up experiments, inadvertent transfer of fabric fibers to dust was minimized via careful vacuuming of the test fabric pre-experiment. Moreover, the organic carbon content of dust was homogeneously distributed (see Table SD-1). Consequently, Figure 2 illustrates the clear increase in concentrations of HBCDs in dust with increasing contact time. This confirms transfer is rapid but that even after 1 week's contact, source:dust equilibrium is not reached. This is consistent with our findings in a recent test chamber study examining the transfer of BDE-209 from plastic TV casing to dust via direct source-to-dust contact (Rauert and Harrad, 2015).

Figure 3 compares HBCD diastereomer profiles in the 4 fabrics studied with those detected in artificial dust samples contacted with the fabrics for 4 days. In each case, there is a demonstrable shift from the pattern in the fabrics, where γ-HBCD predominates, to one where α-HBCD is far more abundant in the dust. This profile shift provides strong evidence that the elevated concentrations in dust in these experiments have not arisen via transfer to dust of abraded fabric fibers. Instead, this pattern shift is consistent with the scenario outlined elsewhere, whereby HBCD uptake by dust occurs via contact with the boundary layer of the source:air interface (Schripp et al, 2010). The reported higher vapor pressure of α-HBCD compared to γ-HBCD (Kuramochi et al, 2010), would yield a boundary layer diastereomer pattern enriched in α-HBCD relative to that in the fabric. In contrast, the similar diastereomer profiles in curtains and post-experiment dust in our test chamber experiments, may be influenced from transferred abraded fabric fibers to the dust. However, our tentative findings of direct HBCD transfer from fabrics to dust, contrast with our earlier findings for PBDE transfer to dust via direct contact with plastic TV casing (Rauert and Harrad, 2015). In this earlier study, we found PBDE congener ratios in dust matched those observed in that of the TV casing, an observation that is more consistent with the hypothesis of Clausen et al (2004), where a dust layer disrupts the boundary layer surrounding the source, allowing direct uptake to dust particles of a chemical from the source surface. Further detailed study of the mechanisms governing BFR transfer from a source to dust therefore appears a research priority to clarify results.

To evaluate the effect of organic carbon content on HBCD transfer from treated fabrics to dust, follow-up experiments were conducted with three different types of

JIS test particles: artificial indoor dust, fine soil particles, and cotton linters. Figure 4 shows that HBCDs in the fabrics transfer to all three dust sample types after 4 days contact time, indicating HBCD transfer is not dominated solely by dust organic carbon content, since the soil particles used contained negligible organic matter. The relative abundance of α-HBCD post-experiment in all the three dust types (Figure 4) exceeded that in the fabrics (Figure 3). Concentrations of HBCDs in dust post-contact Curtain-1, were broadly similar regardless of dust type. In contrast, post-contact Curtain-2, HBCD concentrations in cotton linters were lower than those in other dust types. We cannot explain the different transfer patterns observed for the two curtain samples, but it may be attributable to differences in texture and/or manufacturing process of the two test fabrics.

This study presents substantial evidence that direct contact between HBCD-treated fabrics and dust constitutes an effective transfer mechanism. Comparison with previous test chamber experiments with the same fabric and dust, suggests under real-world conditions, direct source:dust contact may represent the most important transfer pathway for HBCDs. This is consistent with our recent parallel test chamber study in which substantial transfer of PBDEs from plastic TV casing to dust was demonstrated (Rauert and Harrad, 2015). To confirm this novel and important finding, further experiments are recommended to evaluate in more detail the importance of and the mechanisms governing direct source:dust contact as a pathway via which HBCDs and other BFRs undergo transfer from source materials to indoor dust. A particular focus should include studies to elucidate the role of the source:dust boundary layer in effecting contaminant transfer. Our findings have practical implications for reducing

contamination of indoor dust with contaminants like HBCDs, as they imply regular removal of dust from source surfaces will reduce contamination.

Acknowledgements

The research leading to these results received funding from the European Union Seventh Framework Program (*FP7/2007-2013*) under grant agreement No 264600 (INFLAME project), and from the Ministry of Economy, Trade and Industry, Japan.

Dedication

This article is dedicated to the memory of our friend and colleague, Dr. Hidetaka Takigami, who sadly died after completion of this study.

Supplementary Data

Supplementary data to this article consists of descriptions of methods used to determine HBCDs in samples, plus tables reporting QA/QC data and concentrations of HBCDs in follow-up experiment dust samples.

References

Abdallah, M.A.-E., Harrad, S., Covaci, A., 2008a. Hexabromocyclododecanes and Tetrabromobisphenol-A in Indoor Air and Dust in Birmingham, UK: Implications for Human Exposure. Environ. Sci. Technol., 42, 6855-6861.

Abdallah, M.A.-E., Harrad, S., Ibarra, C., Diamond, M.L., Melymuk, L., Robson, M., Covaci, A., 2008b. Hexabromocyclodocanes in indoor dust from Canada, United Kingdom and United States. Environ. Sci. Technol., 42, 459-464. Commercial Use or Development J. Fire Sci. 25, 241-265.

Abdallah, M.A.-E., Tilston, E., Harrad, S., Collins, C., 2012. *In vitro* assessment of the bioaccessibility of brominated flame retardants in indoor dust using a colon extended model of the human gastrointestinal tract. J. Environ. Monit., 14, 3276-3283.

Allen, J.G., Stapleton, H.M., Vallarino, J., McNeely, E., McClean, M.D., Harrad, S.J., Rauert, C.B., Spengler, J.D., 2013. Exposure to flame retardant chemicals on commercial airplanes. Environ. Hlth., 12, 17.

Clausen, P.A., Hansen, V., Gunnarsen, L., Afshari, A., Wolkoff, P., 2004. Emission of Di-2-ethylhexyl Phthalate from PVC Flooring into Air and Uptake in Dust: Emission and Sorption Experiments in FLEC and CLIMPAQ. Environ. Sci. Technol., 38, 2531-2537.

Covaci, A., Gerecke, A.C., Law, R.J., Voorspoels, S., Kohler, M., Heeb, N.V., Leslie, H., Allchin, C.R.; de Boer, J., Hexabromocyclododecanes (HBCDs) in the Environment and Humans: A Review. 2006. Environ. Sci. Technol., 40, 3679-3688.

Harrad, S., Abdallah, M.A.-E., Covaci, A., 2009. Causes of variability in concentrations and diastereomer patterns of hexabromocyclododecanes in indoor dust. Environ. Int., 35, 573-579.

Harrad, S., de Wit, C.A., Abdallah, M.A-E., Bergh, C., Björklund, J.A., Covaci, A., Darnerud, P.O., de Boer, J., Diamond, M., Huber, S., Leonards, P., Mandalakis, M., Östman, C., Småstuen Haug, L., Thomsen, C., Webster, T.F., 2010. Indoor Contamination with Hexabromocyclododecanes, Polybrominated Diphenyl Ethers and Perfluoroalkyl Compounds: An Important Exposure Pathway for People? Environ. Sci. Technol. 44, 3221–3231.

JIS, 2006. Test powders and test particles, Z 8901 (in Japanese)

Kajiwara, N., Desborough, J., Harrad, S., Takigami, H., 2013. Photolysis of brominated flame retardants in textiles exposed to natural sunlight. Environ. Sci. Proc. Imp., 15, 653-660.

Keller, J.M., Stapleton, H.M., Heltsley, R., Peck, A., Kucklick, J.R., Schantz, M.M., Wise, S.A., 2007. Standard reference materials available from the National Institute of Standards and Technology for the analysis of brominated flame retardants. Poster presented at BFR 2007; The Netherlands: Amsterdam.

Kuramochi H., Sakai S., 2010. Measurement of vapor pressures of some PBDEs and HBCD diastereoisomers. Presented at BFR2010, Kyoto, Japan, Abstracts, 90071.pdf

Rauert, C., Lazarov, B., Harrad, S., Covaci, A., Stranger, M., 2014a. A review of chamber experiments for determining specific emission rates and investigating migration pathways of flame retardants. Atmos. Environ., 82, 44-55.

Rauert, C., Harrad, S., Suzuki, G., Takigami, H., Uchida, N., Takata, K., 2014b. Test chamber and forensic microscopy investigation of the transfer of brominated flame retardants into indoor dust via abrasion of source materials. Sci. Tot. Environ., 493, 639-648.

Rauert, C., Harrad, S., 2015. Mass transfer of PBDEs from plastic TV casing to indoor dust via three migration pathways — A test chamber investigation. Sci. Tot. Environ., 536, 568-574.

Rauert, C., Harrad, S., Stranger, M., Lazarov, B., 2015. Test chamber investigation of the volatilization from source materials of brominated flame retardants and their subsequent deposition to indoor dust. Indoor Air, 25, 393-404.

Schripp, T., Fauck, C., Salthammer, T., 2010. Chamber studies on mass-transfer of di(2-ethylhexyl)phthalate (DEHP) and di-n-butylphthalate (DnBP) from emission sources into house dust. Atmos. Environ., 44, 2840-2845.

Takigami, H., Suzuki, G., Hirai, Y., Sakai, S.-I., 2008. Transfer of brominated flame retardants from components into dust inside television cabinets. Chemosphere, 73, 161-169.

Wagner, J., Ghosal, S., Whitehead, T., Metayer, C., 2013. Morphology, spatial distribution, and concentration of flame retardants in consumer products and environmental dusts using scanning electron microscopy and Raman micro-spectroscopy. Environ. Int., 59, 16-26.

Webster, T.F., Harrad, S., Millette, J.R., Holbrook, R.D., Davis, J.M., Stapleton, H.M., Allen, J.G., McClean, M.D., Ibarra, C., Abdallah, M.A.-E., Covaci, A., 2009. Identifying Transfer Mechanisms and Sources of Decabromodiphenyl Ether (BDE 209) in Indoor Environments Using Environmental Forensic Microscopy, Environ. Sci. Technol., 43, 3067-3072.

Weil, E.D., Levchik, S.V., 2007. Flame Retardants for Polystyrenes in Suzuki, G., Kida, A., Sakai, S., Takigami, H., 2009. Existence state of bromine as an indicator of the source of brominated flame retardants in indoor dust. Environ. Sci. Technol. 43, 1437–1442.

Figures and Tables

Figure 1: Schematic of experimental configurations used in: (a) test chamber (top) and (b) follow-up experiments (bottom) investigating uptake via direct contact between source and dust

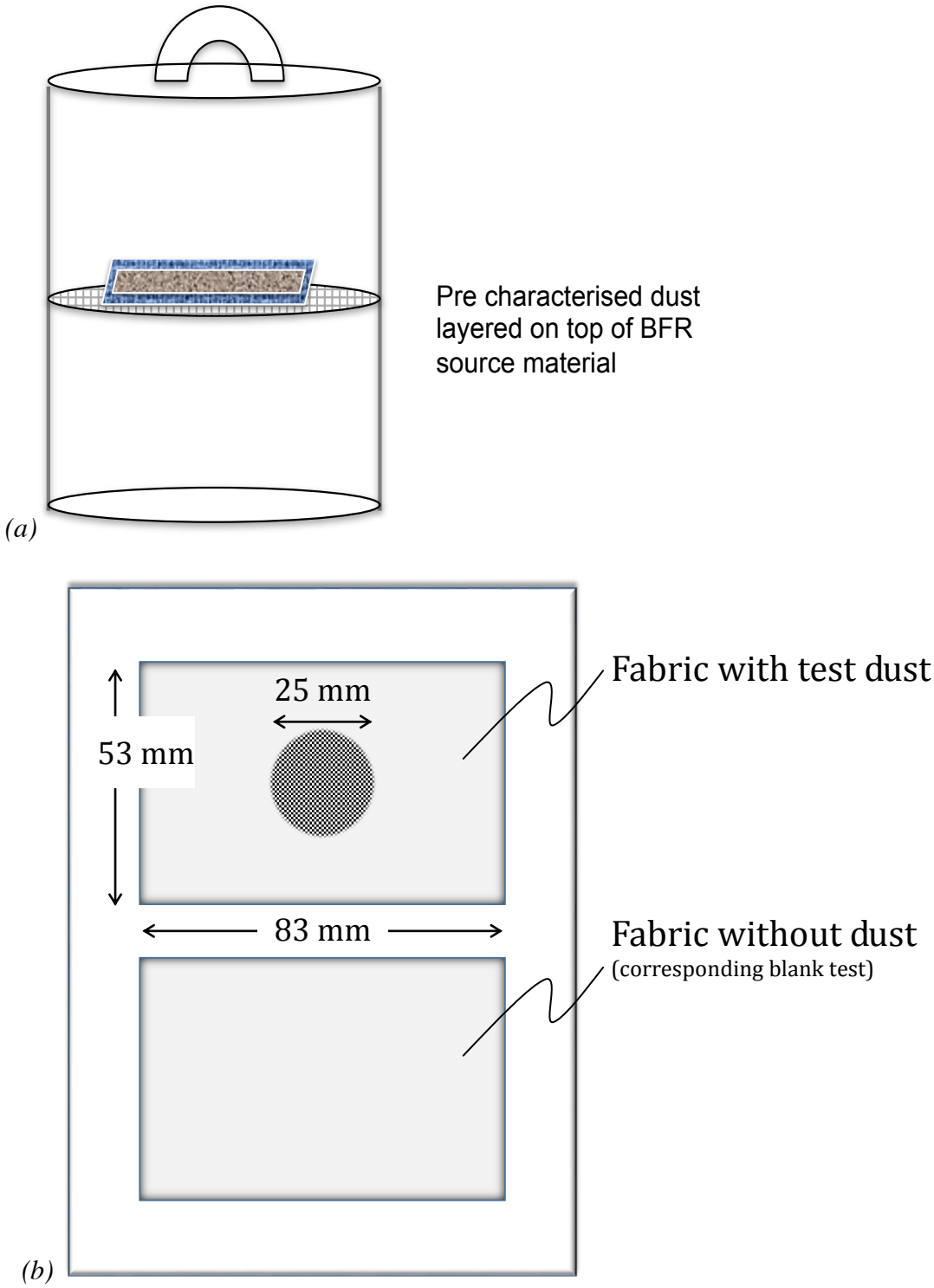
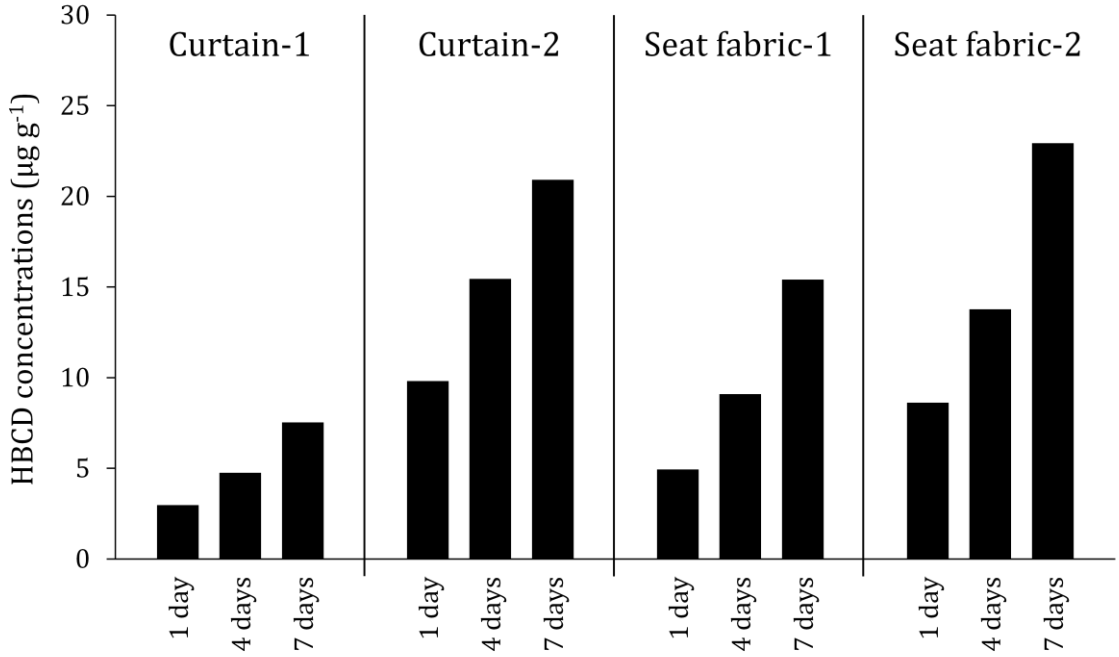


Figure 2. Temporal changes in ΣHBCD concentrations (*μ*g/g)* in JIS artificial dust after the follow-up experiments.



*After subtracting HBCD concentrations detected in the corresponding blank test. Values for Curtain-1 and Seat fabric-1 are the average of duplicate analyses.

Figure 3. HBCD diastereomer profiles in the original fabrics and the artificial dust samples placed in direct contact with the fabrics in the follow-up experiments.

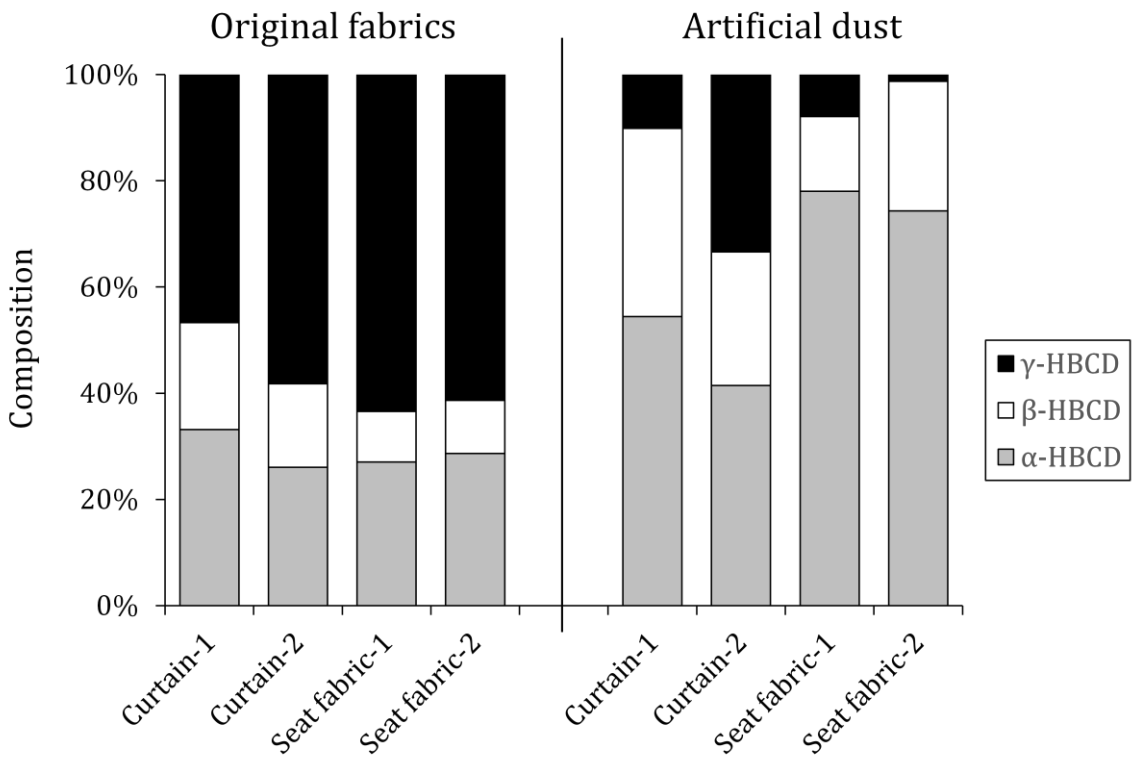


Figure 4. Differences in HBCD diastereomer concentrations transferred to the three different types of JIS test particles during 4 days of contact with treated curtains.

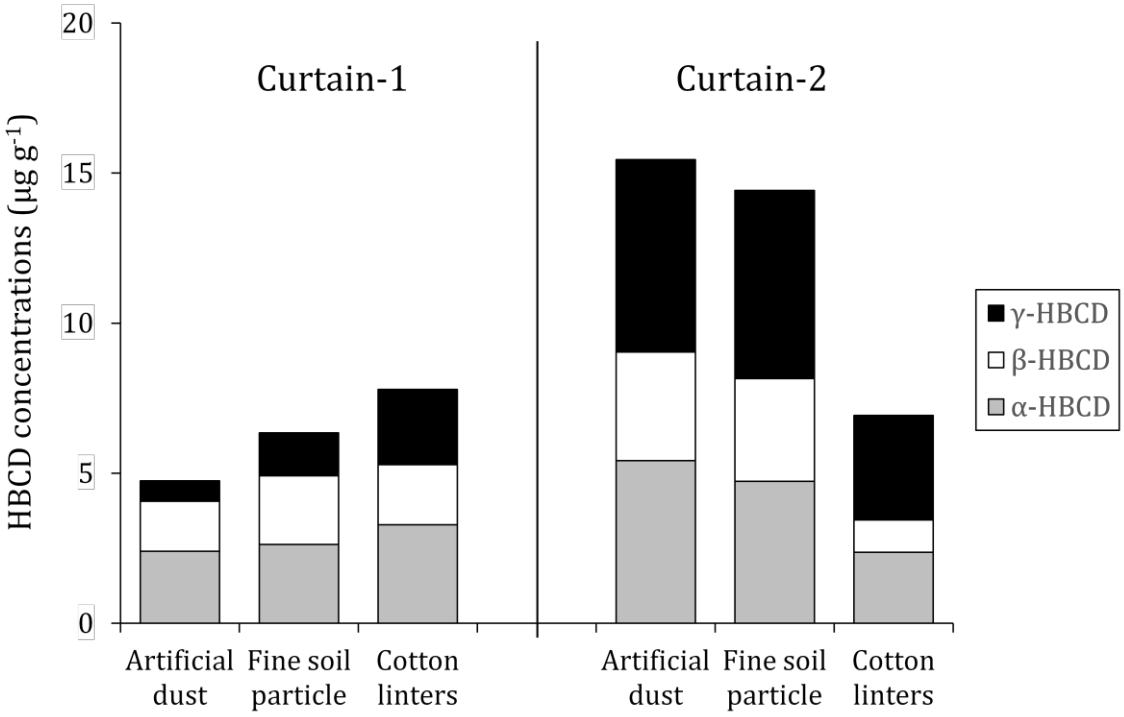


Table 1: Mean and (min, max) concentrations (ng g^{-1}) of HBCDs in dust from direct contact test chamber experiments conducted for 24 hours and 1 week

		α-HBCD	β-HBCD	γ-HBCD	ΣHBCDs
Pre experiment dust (ng g^{-1})		*46 ± 18*	*13 ± 10*	*50 ± 39*	*110*
24 hours	Experiment 1 (n=2)	600 (580, 630)	200 (190, 210)	730 (730, 740)	1500 (1500, 1600)
	Experiment 2 (n=2)	1800 (1500, 2000)	530 (430, 630)	2000 (1700, 2400)	4300 (3600, 5000)
	Experiment 3 (n=2)	5400 (2600, 8200)	1700 (860, 2500)	6400 (3500, 9400)	14 000 (6900, 20 000)
	Experiment 4 (n=2)	2500 (2200, 2700)	800 (750, 860)	3200 (2900, 3500)	6500 (5900, 7000)
1 week	Experiment 1 (n=2)	3700 (3600, 3800)	1300 (1200, 1400)	4300 (4000, 4600)	9300 (8800, 9800)
	Experiment 2 *(n=1)*	3000	910	3400	7300
	Experiment 3 (n=2)	3000 (1900, 4100)	920 (500, 1400)	3500 (2000, 4900)	7400 (4400, 10 000)
	Experiment 4 (n=2)	2300 (2100, 2400)	590 (560, 620)	2300 (2100, 2600)	5200 (4700, 5600)

Table 2: Mean ± standard deviation concentrations (ng g^{-1}) of HBCDs in dust post-test chamber experiments simulating three different migration pathways of HBCDs to dust from HBCD treated curtains

	α-HBCD	β-HBCD	γ-HBCD
Volatilization with subsequent partitioning to dust[a]			
24 hours @ 60 °C (n=4)	180 ± 52	63 ± 3.8	370 ± 85
1 week @ 22 ± 1 °C (n=3)	160 ± 100	72 ± 60	270 ± 180
Abrasion of fibers directly to dust[a]			
2 hours (n=1)	2500	720	3300
3 hours (n=1)	1400	700	2000
21 hours (n=1)	3200	1700	13 000
48 hours (n=1)	23 000	4900	26 000
Direct transfer from source to dust (this study)			
24 hours (n=4)	2600 ± 2000	800 ± 620	2900 ± 2400
1 week (n=4)	3000 ± 600	930 ± 280	3400 ± 790

[a]Data reported in Rauert et al (2014b)

Table 3: Initial concentrations (mg/kg) of HBCD diastereomers in the fabric used in the follow-up experiments

ID	Material	α-HBCD	β-HBCD	γ-HBCD	ΣHBCDs
Curtain-1	polyester	7,100	4,300	10,000	21,000
Curtain-2	polyester	3,800	2,300	8,500	15,000
Seat fabric-1	polyester	2,300	810	5,400	8,400
Seat fabric-2	polyester	4,300	1,500	9,200	15,000

Table 4: HBCD concentrations (μg/g)* in JIS test dust after the follow-up experiments

JIS test dust	Test fabric	Experimental duration (days)	α-HBCD	β-HBCD	γ-HBCD	ΣHBCDs
Artificial dust	Curtain-1**	1	1.4	1.1	0.53	3.0
		4	2.4	1.7	0.68	4.7
		7	4.1	2.7	0.76	7.5
	Curtain-2	1	3.4	2.1	4.3	9.8
		4	5.4	3.6	6.4	15
		7	8.7	5.3	7.0	21
	Seat fabric-1**	1	3.7	0.70	0.50	4.9
		4	7.1	1.3	0.69	9.1
		7	12	2.2	1.2	15
	Seat fabric-2	1	6.3	2.2	0.18	8.6
		4	10	3.4	0.13	14
		7	17	5.6	0.29	23
Cotton linter	Curtain-1**	4	3.3	2.0	2.5	7.8
	Curtain-2	4	2.4	1.1	3.5	6.9
Fine soil particle	Curtain-1**	4	2.6	2.3	1.4	6.3
	Curtain-2	4	4.7	3.4	6.3	14

* After subtracting HBCD concentrations in the corresponding blank test.
** Values are averaged from duplicate analyses.

Supplementary material for on-line publication only

Click here to download Supplementary material for on-line publication only: Rauert et al 2015 SD R2.docx